Effective cavity pumping from weakly coupled quantum dots

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Abstract

We derive the effective cavity pumping and decay rates for the master equation of a quantum dot-microcavity system in presence of N weakly coupled dots. We show that the in-flow of photons is not linked to the out-flow by thermal equilibrium relationships.

Keywords: quantum dots, microcavities, strong coupling, PL spectrum
PACS: 42.50.Ct, 78.67.Hc, 42.55.Sa, 32.70.Jz

Strong coupling between a single quantum dot (QD) an a microcavity mode was achieved for the first time in 2004 [1, 2] and has since witnessed many technical progresses, from hitting the quantum limit [3] and quantum nonlinearities [4] to lasing [5] and scalable implementation [6]. Prospects for this field are great given the ever increasing quality of structures and reaching better strong coupling (see [7] for a review). On the theoretical side of this fundamental physics, we have shown that including the incoherent continuous excitation, typical of these experiments, is essential to reproduce quantitatively the observed spectral anticrossing [8]. Non-vanishing pumping also affects the structure of dressed states [9, 10] as compared to its spontaneous emission counterpart. The direct and continuous excitonic pumping is provided by the income of electron-hole pairs optically generated or electrically injected in the wetting layer [11]. However, also some cavity pumping is necessary in order to successfully fit the spectral lineshapes [8, 12, 13]. Different mechanisms may produce some income of cavity photons, such as the well known thermal excitation [14] or the cascade de-excitation of multiexciton states [15]. It has been recently argued that only the thermal type of excitation is physically admissible for the cavity [16, 17]. This was refuted in Ref. [18] based on physical arguments. Here, we show that a simple model can make explicit a case where cavity pumping is not, indeed, of a thermal character. The model describes the situation—appealing on physical grounds—where the dot that strongly couples to the cavity mode, is surrounded by several "spectator" dots that are in weak-coupling. These also get excited by the excitonic pumping, and emit preferentially, due to Purcell enhancement, in the cavity mode.

We thus consider an assembly of QDs with Fermion lowering operators σ_i (i labelling the ith dot), each coupled with strength g_i to the single Boson mode a of a microcavity. The QDs are detuned by a small quantity Δ_i from the cavity mode, with the rotating wave Hamiltonian: $H_i = \Delta_i\sigma_i^\dagger\sigma_i + g_i(a^\dagger\sigma_i + a\sigma_i^\dagger)$. Each dot is further

endowed with dissipation (at rate γ_i) and excitonic pumping (at rate P_i) in the Lindblad form, $\mathcal{L}_i\rho = \frac{\gamma_i}{2}(2\sigma_i\rho\sigma_i^\dagger - \sigma_i^\dagger\sigma_i\rho - \rho\sigma_i^\dagger\sigma_i) + \frac{P_i}{2}(2\sigma_i^\dagger\rho\sigma_i - \sigma_i\sigma_i^\dagger i\rho - i\rho\sigma_i\sigma_i^\dagger)$. The cavity dissipation (at rate γ_a) and cavity pumping (at rate P_a) also are in the Lindblad form, $\mathcal{L}_a\rho = \frac{\gamma_a}{2}(2a\rho a^\dagger - a^\dagger a\rho - \rho a^\dagger a) + \frac{P_a}{2}(2a^\dagger\rho a - aa^\dagger\rho - \rho aa^\dagger)$. The total density matrix operator ρ, follows the master equation:

$$\partial_t\rho = \sum_i \Big(i[\rho, H_i] + \mathcal{L}_i\rho\Big) + \mathcal{L}_a\rho\,. \tag{1}$$

If the dots are uncoupled to the cavity, $g_i = 0$, we can easily obtain from the rate equations $\partial_t\langle a^\dagger a\rangle = -\gamma_a\langle a^\dagger a\rangle + P_a(1+\langle a^\dagger a\rangle)$ and $\partial_t\langle\sigma_i^\dagger\sigma_i\rangle = -\gamma_i\langle\sigma_i^\dagger\sigma_i\rangle + P_i(1-\langle\sigma_i^\dagger\sigma_i\rangle)$, the steady state populations for cavity mode and dots:

$$n_a \equiv \langle a^\dagger a\rangle = P_a/\Gamma_a\,, \quad n_i \equiv \langle\sigma_i^\dagger\sigma_i\rangle = P_i/\Gamma_i\,. \tag{2}$$

They are given in terms of the bosonic and fermionic effective decoherence rates:

$$\Gamma_a \equiv \gamma_a - P_a\,, \quad \Gamma_i \equiv \gamma_i + P_i\,, \tag{3}$$

which are also the uncoupled spectral linewidths of cavity and dots, as obtained through the quantum regression formula [19]. The number of photons can be arbitrarily large while each QD takes (average) values between 0 and 1. If the coupling is weak or the detuning to the cavity mode is large, the populations and linewidths are still given by expressions (2) and (3), but for some effective decay and pump parameters, γ^{eff}, P^{eff}, as shown below.

All Γs and Ps are as yet essentially undefined phenomenological parameters. They could be linked in some way, e.g., if the dissipation arises from a thermal bath, the following well known relationship would link them (in both cavity and QD cases):

$$\gamma = \kappa(\bar{n}_T + 1)\,, \quad P = \kappa\bar{n}_T\,. \tag{4}$$

κ is the zero temperature decay rate of the mode and $\bar{n}_T$ is the mean number of excitations in the reservoir at temperature T. This leads to thermal equilibrium average populations $n_a = \bar{n}_T$ and $n_i = \bar{n}_T/(2\bar{n}_T + 1)$ and naturally prohibits features of out-of-equilibrium, such as inversion of population for the dot (here n_i remains below $1/2$) or line narrowing for the cavity ($\Gamma_a = \kappa_a$ remains a constant, independent of T), however high the occupancy $\bar{n}_T$ of the thermal mode. A thermal bath is a medium of *loss*, as $\gamma > P$ (as seen clearly in Eq. (4)). In out-of-equilibrium conditions, especially under an externally applied pumping, one can expect deviations from the thermal scenario. We will see in what follows a simple model of a *gain* medium for the cavity, in the sense that the linewidth decreases while the effective pumping rate increases, with the external excitation.

In the bosonic case, if P_a and γ_a are allowed to vary independently, there is an obvious singularity in Eq. (3) at $P_a = \gamma_a$, past which point values are negative. This is because, although the master equation is still valid at all finite time, it has no steady state. The physical reason why, is clear enough: more particles are injected at all time than are lost by decay. Therefore populations increase without bound (they diverge

in the infinite times). There is no deeper physics here than the fact that no all dynamical systems have a steady state, some because they are oscillatory, others because they increase without bounds. The general consideration of pump and decay bosonic rates, finds its most important domain of applicability with atom lasers and polariton lasers [20, 21, 22, 23, 24, 25, 26, 27], that is, systems where a condensate (or coherent state) is formed by scattering of bosons into the final state from another state rather than by emission. In both cases, scattering or emission, the process is stimulated. In this case the income and outcome of particles is a complicated function of the distribution of excitons (or polaritons) in the higher k-states (see, e.g., Ref [21]) and even the case $\gamma_a(t) < P_a(t)$ can then be realized in the transients [28].

On the other hand, in the fermionic (QD) case, there are no divergences, and parameters P_i and γ_i are, in general, considered as independent in the literature. In fact, within the theory of the single-atom laser, a more general relationship between the emitter pump and decay rates, is broadly used [29]: $\gamma_i = \Gamma_i(1 - s_i), \quad P_i = \Gamma_i s_i$, where s_i is limited to the interval $[0, 1]$. This form now describes when $s_i > 1/2$, *gain* of the QD from the reservoir which leads to its population inversion $n_i > 1/2$. This can be theoretically mapped to a thermal bath with negative temperature [30].

We consider that among all the dots in the sample, only one, say $i = 0$, couples strongly ($g_0 \gg \gamma_a$) and resonantly ($\Delta_0 = 0$) to the cavity mode. The dynamics of strong coupling between this dot and the cavity, the so-called *Jaynes-Cummings* model, cannot be described perturbatively and must in general be solved to all orders and numerically [9]. We further investigate the rest of the dots in the sample, $i = 1, \dots N$, that are weakly coupled or/and far from resonance to the cavity mode. Their effect on the cavity population and spectral properties is perturbative (Purcell effect) and can be considered, as a first approximation, independent of the strong coupling physics with dot 0. We, therefore, solve Eq. (1) with $i > 0$, to first order and then trace out the weakly coupled dots degrees of freedom. This will result in some effective parameters γ_a^{eff} and P_a^{eff} that are to appear eventually in the final reduced master equation, that was the starting point in our earlier work:

$$\partial_t\rho = i[\rho, H_0] + \mathcal{L}_0\rho + \frac{\gamma_a^{\mathrm{eff}}}{2}(2a\rho a^\dagger - a^\dagger a\rho - \rho a^\dagger a) + \frac{P_a^{\mathrm{eff}}}{2}(2a^\dagger\rho a - aa^\dagger\rho - \rho aa^\dagger)\,. \quad (5)$$

The line broadening of the cavity mode neglecting the strongly-coupled dot (when $g_0 = 0$) is given by $\Gamma_a^{\mathrm{eff}} = \gamma_a^{\mathrm{eff}} - P_a^{\mathrm{eff}}$.

We start by solving the dynamics of the cavity with only one of the weakly coupled dots: $i = 1$. Only one-photon correlations need be considered, which is equivalent to solving the dynamics truncating in the first rung [31, 9]. The solutions are analytical, $n_a = P_a^{\mathrm{eff}}/\Gamma_a^{\mathrm{eff}}$, $n_1 = P_1^{\mathrm{eff}}/\Gamma_1^{\mathrm{eff}}$, in terms of the effective pumping rate and line broadenings $P_a^{\mathrm{eff}} = P_a + \frac{Q_{a1}}{\Gamma_a+\Gamma_1}(P_a + P_1)$, $\Gamma_a^{\mathrm{eff}} = \Gamma_a + Q_{a1}$, $P_1^{\mathrm{eff}} = P_1 + \frac{Q_1}{\Gamma_a+\Gamma_1}(P_a + P_1)$, $\Gamma_1^{\mathrm{eff}} = \Gamma_1 + Q_1$ and the Purcell exchange rate of the cavity into the dot, $Q_{a1} = 4(g_1^{\mathrm{eff}})^2/\Gamma_1$, and the dot into the cavity mode, $Q_1 = 4(g_1^{\mathrm{eff}})^2/\Gamma_a$. The effective coupling strength appearing in these expressions is given by $g_1^{\mathrm{eff}} = g_1/\sqrt{1 + \left(\frac{2\Delta_1}{\Gamma_a+\Gamma_1}\right)^2}$. By solving the system with two, three, etc., weakly coupled dots, we find the general cavity effective parameters

for N dots:

$$P_a^{\text{eff}} = P_a + \sum_{i=1}^{N} \frac{Q_{ai}}{\Gamma_i + \Gamma_a}(P_i + P_a) + \sum_{\{i,j\}} \frac{Q_{ai}Q_{aj}}{(\Gamma_a + \Gamma_i)(\Gamma_a + \Gamma_j)}(P_i + P_j + P_a) + \dots\,, \quad (6a)$$

$$\Gamma_a^{\text{eff}} = \Gamma_a + \sum_{i=1}^{N} Q_{ai} + \sum_{\{i,j\}} \frac{Q_{ai}Q_{aj}}{(\Gamma_a + \Gamma_i)(\Gamma_a + \Gamma_j)}(\Gamma_a + \Gamma_i + \Gamma_j) + \dots\,. \quad (6b)$$

The sums are taken for increasingly large combinations of dots, $\{i, j, k, \dots\}$, with $i < j < k < \dots$. They correspond to the exchange of a photon between different dots. The larger the group, the smaller its contribution to the effective parameters, as the weight is given by the product $\alpha_i\alpha_j\alpha_k\dots$ where $\alpha_i \equiv Q_{ai}/(\Gamma_a+\Gamma_i)$ is a dimensionless quantity, that is small in the present model.

For simplicity, we consider that all QDs ($i = 1, \dots, N$) are coupled to the cavity mode with similar coupling strength, $g_i = g$, detunings, $\Delta_i = \Delta$, that they have similar decay rates into the leaky modes, $\gamma_i = \gamma$ and are excited at the same pumping rates $P_i = P$ (then, $\Gamma_i = \Gamma$, $g_i^{\text{eff}} = g^{\text{eff}}$, $Q_{ai} = Q_a$ and $\alpha_i = \alpha$). More realistically, there would be a Gaussian distribution of these parameters, however, QDs with higher effective coupling lead the dynamics, and this approximation actually results in little loss of generality. With this simplification, we obtain the compact expressions:

$$P_a^{\text{eff}} = P_a + \sum_{n=1}^{N} \frac{N!\alpha^n}{(N-n)!n!}(P_a + nP) = (P_a + N\frac{\alpha}{1+\alpha}P)(1+\alpha)^N\,, \quad (7a)$$

$$\Gamma_a^{\text{eff}} = \Gamma_a + \sum_{n=1}^{N} \frac{N!\alpha^n}{(N-n)!n!}(\Gamma_a + n\Gamma) = (\Gamma_a + N\frac{\alpha}{1+\alpha}\Gamma)(1+\alpha)^N\,, \quad (7b)$$

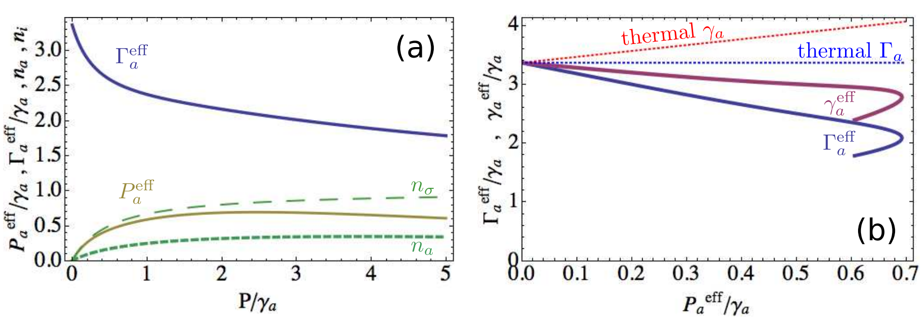

Figure 1: (Colour online) (a) Effective pumping rate, P_a^{eff} (solid brown) and cavity linewidth, Γ_a^{eff} (solid blue) as a function of the pumping rate of the dots, P. In dashed green, the dot populations, n_σ, quickly saturates to 1. In dotted green, the resulting cavity population, n_a. We checked that both populations agree with the Jaynes-Cummings numerical solution at higher orders. (b) Γ_a^{eff} (solid blue) decreases and γ_a^{eff} (solid purple) slightly decreases as a function of P_a^{eff} in contrast with a thermal excitation of the cavity where Γ_a (dotted blue) remains constant and γ_a (dotted red) increases linearly as a function of the same P_a. Note in (a) that P_a^{eff} decreases from $P \approx 2.5$, which provokes the loop in (b) at $P_a^{\text{eff}} \approx 0.7$. Parameters in this example are $N = 15$, $g = 0.3$, $\Delta = 2$, $\gamma = 0.5$, $P_a = 0$. All quantities are in units of γ_a.

For illustration, let us assume that the system is at zero temperature and that the cavity is not excited directly, i.e., $P_a = 0$. Then, P_a^{eff} is fully produced by the Purcell emission of all the weakly coupled QDs. In Fig. 1, we plot the effective cavity parameters as a function of the external QD pumping P (for some parameters given in the caption, typical of the experiments). All magnitudes are given in terms of the empty cavity decay rate, γ_a. We can see in (a) that for $P < 2.5$, P_a^{eff} increases while Γ_a^{eff} monotonically decreases. If we plot one as function of the other, (b), we find line narrowing with an increasing effective cavity pumping. This is to be compared with a thermal excitation (dotted lines), where the linewidth indeed does not change. The effective decay rate γ_a^{eff} also decreases slightly, while it would increase linearly with the cavity pumping in the thermal case.

It is not essential to invert the QD population (see n_σ in (a)) in order to obtain line narrowing. Similar results, with QD saturation into $1/2$ instead of 1, are obtained with a thermal excitation of the QDs ($\gamma = \kappa + P$ and $\kappa = 0.5$). The cavity population remains quite low in any case (see n_a in (a)), as expected from weak-coupling, but this small contribution is enough to cause qualitative differences in the strong coupling physics that involve the QD of interest, $i = 0$ [9].

This simple model has room for arbitrary sophistication that can relate Γ_a^{eff} and P_a^{eff} in Eq. (5) in all possible conceivable ways. It is thus a shortcoming, in a configuration where the simplest model displays opposite tendencies, to assume thermal relationships between Γs and Ps [16, 17], or, for that matter, any particular constrain, such as Eqs. (7). A more general view should be adopted to let these parameters completely free and, based on statistical inference, to extract correlations from them *a posteriori*.

In conclusion, we have derived from a microscopic model of N weakly coupled and incoherently excited quantum dots, the effective cavity pumping and decay rates for a master equation in the Lindblad form. These are not linked by relationships of thermal equilibrium. The QDs emit cavity photons via Purcell enhancement, providing a gain medium for the cavity. As a result, the cavity spectral lineshape narrows with increasing excitation, in contrast with a thermal photonic excitation, where it remains constant.

Acknowledgements: EdV acknowledges the support of a Newton Fellowship.

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